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COMBUSTION AND FLAME PROPAGATION IN HETEROGENEOUS SYSTEMS

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Madrid, Spain

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ABSTRACT

In this report first are summarized the works carried out on unsteady effects in droplet evaporation and combustion and on the behaviour of hydrazine catalytic thrusters working under steady state conditions. The research performed in these two fields was presented in more detail in previous scientific reports.

The third topic, that is presented in more detail, is on the influence of the temperatures of liquid hydrazine and catalyst particle temperatures on the vacuum start characteristics of a reactor. The model used assumes the existence of a liquid front that first advances towards the interior of the catalyst particle by porous action and then recedes towards the exterior. An order of magnitude estimate seems to indicate that during the process, convective heat transfer dominates over conduction, and the analysis shows the existence of a thin thermal front, whose structure is calculated. Then, the liquid penetration is only governed by the initial particle temperature. For low enough initial particle temperatures the thermal front structure reaches the liquid front and the liquid penetration is also influenced by the initial hydrazine temperature.

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1. INTRODUCCION AND SUMMARY OF PREVIOUS RESEARCH WORK

During the report period work has been done on the following three areas.

a) Unsteady Effects in Droplet Evaporation and Combustion.

b) Steady State Analysis of Hydrazine Catalytic Thrusters.

c) Influence of both Liquid Hydrazine and Catalyst Particle Temperatures on the Vacuum Start Characteristics of a Reactor.

The work carried out on the first two subjects has already been presented in detail in three previous Scientific Reports and only will be summarized in the present report. Work on topic c) has been done mainly during this year and will be presented in more detail.

1.1 Unsteady Effects in Droplet Evaporation and Combustion

The work carried out on this subject has been presented already in a report by Crespo and Liñan¹ this has also appeared with some modifications as a journal article (see also ref.1).

The problem considered was that of the droplet vaporization and combustion in a stagnant atmosphere taking into account the unsteady effects in the gas phase. Previous works on the subject assumed the process to be quasisteady, based on the fact that the ratio

of the densities of the ambient gas and of the liquid was small. In this work a perturbation analysis is made in which, by considering the above mentioned ratio as small, the asymptotic nature of the quasisteady (QS) solutions is shown. The analysis shows that corrections to the QS theory are of the order of the square root of the ratio of the densities and that the solution is non-steady (NS) at distances from the droplet of the order of the droplet radius divided by the square root of the ratio of the densities.

It is found that the droplet lifetime is given by the simple analytical expression

$$\frac{t_1}{t_{10}} = 1 / (1 + \sqrt{2\epsilon m_0 / \pi}) \quad (1)$$

Where t_{10} is the lifetime that would be obtained from the QS analysis, ϵ is the abovementioned ratio of the densities of gas and liquid fuel, and m_0 is a parameter defined in ref. 1 and that is inversely proportional to t_{10} . A comparison is given in figure 1 of the result given by expression (1) with those obtained numerically by Duda and Vrentas² for the problem of dissolution of spheres, that can be shown to be mathematically similar to this one. The agreement is good, even for values of ϵm_0 of order unity, for which the present perturbation theory can not be applied.

For the combustion case the flame position and extinction time have also been investigated. When the

stoichiometric ratio oxidizer fuel is not very large it is found that QS theories give flame position correctly with a relative error of order $\sqrt{\epsilon}$. In many practical cases this ratio is large, even of the order of $\epsilon^{-1/2}$, and the flame is located then in the NS region and its position is not given, even in first approximation, by the QS theory. Comparisons have been carried out with experimental results of Kumagai et al³ that show a reasonable agreement. Also when the stoichiometric ratio is large, it is found that the droplet lifetime is shorter than the flame extinction time, and an analytical expression is presented giving this extinction time. For cases in which the extinction time is much longer than the lifetime the obtained results coincide with those obtained by Spalding⁴ who replaced the droplet by an instantaneous source of vapor.

1.2 Steady State Analysis of Hydrazine Catalytic Thrusters

The results obtained in this area have been presented in detail in two previous reports by A. Crespo, E. Fraga and A. Muñoz⁵ and A. Crespo⁶. In reference 5 the problem is stated and some guidelines for the solution are given. The most important results appear in ref.6.

The model examined is a onedimensional stationary one in which liquid hydrazine enters the reactor and decomposes yielding enough heat to vaporize it and heat the mixture. The hydrazine decomposition process is assumed to follow a two-step reaction mechanism in which hydrazine decomposes exothermically yielding ammonia,

hydrogen and nitrogen, and the ammonia in turn decomposes endothermically producing hydrogen and nitrogen. The purpose of the work is to find simple expressions for the temperature and mass fraction distributions along the chamber and in particular at the exit as a function of the operating conditions. From these exit conditions the specific impulse that would be obtained if the mixture would be expanded to the vacuum is obtained.

Extensive numerical solutions of the problem are given by Kesten⁷ in which a large number of parameters should be used as input for his computer program although he also gives a numerical correlation of his results. In a work carried out previously at INTA⁸ also numerical solutions were obtained but the number of parameters was practically reduced to one, although the results were only applicable for Shell 405 Catalyst with established values of the velocity and stoichiometry of the reaction.

In the work carried out with the present Grant some simplifications were introduced to the model of ref.8 the most important one being to consider that the ammonia decomposition is only important at temperatures close to the maximum one that is reached in the reactor. By means of this simplification an analytical solution is obtained that gives conditions at the chamber exit, in particular specific impulse for mixture expanded to the vacuum and fractional ammonia decomposition, as functions of operating conditions and chamber characteristics for different types of catalyst particles and chemical kinetics. These results show a reasonable agreement with those of Kesten⁷, and as a matter of fact our analytical expression for fractional ammonia decomposition reproduces some aspects of his correlation of numerical results.

1.3 Separate influence of catalyst and liquid hydrazine temperatures on vacuum start characteristics of a hydrazine catalytic reactor.

During the last part of the working period corresponding to this grant, work has been done on studying the temperature distribution within a catalyst particle during the period of liquid hydrazine penetration inside this particle, when the initial temperatures of hydrazine and catalyst are different.

One of the most important problem areas in the field of hydrazine catalytic decomposition for satellite microthrusters, is the catalyst attrition originated by the large overpressures created inside the particles during long ignition delays. This ignition delay time is strongly dependent on the initial temperature at which this ignition takes place. It is the purpose of our work to study the separate influence of both the initial temperatures of catalyst and liquid hydrazine on the ignition delay.

The model used for the study is similar to those used by Sangiovanni and Kesten⁹ and in a previous work at INTA⁸. Hydrazine penetrates inside the catalyst by porous action and a liquid front advances towards the interior of the particle; at the interphase hydrazine vaporizes, and these vapours, in contact with the active sites in the walls decompose catalitically. As the liquid front advances, and more hydrazine is decomposed, the gas pressure increases until finally it overcomes the capillary pressure and the liquid front starts moving

towards the particle surface. A different model has been proposed by Webber and Schechter¹⁰ in which there is not definite liquid front, but gaseous hydrazine and products are allowed to coexist with liquid hydrazine throughout the catalyst particle; although this may be a more realistic model than the one used here, we shall not use it since it is thought that a physical insight of the process can be obtained without recurring to this more complicated model.

In all the abovementioned works the calculations were performed for equal or nearly equal initial temperatures of liquid hydrazine and catalyst.

The calculations carried out in this report show the existence of a thermal front moving behind the liquid front. Ahead of the thermal front the liquid hydrazine is at the initial temperature of the catalyst, T_0 , and behind at the initial temperature of the liquid, T_1 (figure 2). The internal structure of this thermal front has been calculated. The reason for the existence of the front lies in the fact that convective heat transfer is more important than heat conduction; this has also been observed by Webber and Schechter¹⁰. Since the evaporation and reaction of hydrazine takes place at the liquid front, in the region inside it, the results of the report would indicate that the only temperature that would have an influence on the ignition delay would be the initial particle temperature and not the liquid hydrazine temperature. Some experiments carried out by Fouche et al.¹¹ confirm that the ignition time is independent of liquid hydrazine temperature if the catalyst bed temperature is above a certain critical value, however for smaller values of the catalyst tem-

perature the ignition time becomes dependent on both liquid and catalyst temperatures, thus invalidating our theory in this second situation. Our interpretation is that for low temperatures the ignition time, and hence the residence time of the liquid inside the particle, becomes long, thus decreasing the importance of the convective effects that become of the same order of the heat conduction. The limiting conditions under which this occurs will also be investigated.

Other experiments on this area have been carried out by Sangiovanni and Kesten¹² and Martney and Kesten,¹³ however they are difficult to compare with our theory because they either use the same temperature for liquid hydrazine and catalyst or do not state clearly the difference between both of them.

We shall now describe in more detail this problem in the following pages.

2. TEMPERATURE DISTRIBUTION WITHIN A CATALYST PARTICLE DURING PENETRATION OF LIQUID HYDRAZINE

The problem to be studied consists in the calculation of the temperature distribution within a porous spherical particle that is initially at a temperature T_0 when a liquid at a temperature T_1 penetrates in it a prescribed flow rate Q that is a function of time. The main assumptions made in the following are as follows:

- a) Radial symmetry
- b) Liquid hydrazine penetrates in the particle at a prescribed rate $Q(t)$ that we suppose to be known. We shall explain later how this datum can be obtained from previous calculations made assuming that $T_1 = T_0$, (refs. 8,9)
- c) All the pores between the liquid front radius R and the particle radius, a , are filled with liquid.
- d) We neglect the vaporization rate of hydrazine compared with its mass flow rate during this initial unsteady process. This may be justified by comparing typical values of the masses of gas and liquid in the particle or of their densities if the volumes they occupy are comparable; at a typical gas pressure of 100 atm. the gas density is an order of magnitude smaller than the liquid density. Under this assumption Q and R will be related by:

$$\gamma \frac{4}{3} \pi (a^3 - R^3) = \int_0^t Q(t) dt \quad (2)$$

γ being the volume fraction occupied by the pores.

- e) At a given time and location within the particle the fluid and solid temperature are equal.
- f) There is an overall thermal conductivity, K_p , that is supposed to be temperature independent.
- g) Temperature changes due to other causes besides the unequal initial temperatures of liquid and particle are neglected. Sangiovanni and Kesten found in their numerical calculations temperature rises of less than 4°K for cases in which these initial temperatures were equal.

With these assumptions the equation governing heat transfer within the particle in the liquid region, $R < r < a$, is

$$\rho c \frac{\partial T}{\partial t} - \frac{Q \rho_1 c_1}{4\pi r^2} \frac{\partial T}{\partial r} = \frac{1}{r^2} \frac{\partial}{\partial r} (K_p r^2 \frac{\partial T}{\partial r}) \quad (3)$$

ρc is an average value of specific heat per unit volume

$$\rho c = \rho_s c_s (1-\gamma) + \rho_1 c_1 \gamma \quad (4)$$

ρ_s and ρ_1 are the densities of the solid forming the catalyst and of the liquid hydrazine, and c_s and c_1 the corresponding specific heats per unit mass.

2.1 First Approximation

It is obtained by neglecting the last term of equation (3) representing heat conduction. By comparing this term with the second one representing heat convection, it is obtained that their ratio is of order of the ratio of the liquid residence time within the par-

ticle, t_r , and the characteristic time for heating of the particle by conduction, t_c

$$t_r = \frac{4/3\pi a^3}{Q_0} \quad t_c = \frac{\rho_1 c_1 a^2}{K_p} \quad (5)$$

Where Q_0 is a typical value of Q . For particles of radius of the order of .5mm., refs. 8,9 give for t_r values below 0.1 sec. for temperatures above 290°K. A typical value of t_c for that particle radius is 1 sec.

Equation (3) without the conduction term becomes of the hyperbolic type and has a general solution of the form

$$T=f(\xi) \quad (6)$$

$$\text{Where } \xi = \frac{4}{3} \pi (a^3 - R^3) - \frac{\rho c}{\gamma \rho_1 c_1} \frac{4}{3} \pi (a^3 - r^3) \quad (7)$$

and equation (2) has been used.

The initial and boundary conditions to be satisfied are

$$t=0, \quad r < a, \quad T=T_0 \quad (8)$$

$$t \geq 0, \quad r=a, \quad T=T_1 \quad (9)$$

With these conditions $f(\xi)$ becomes a step function such that

$$f(\xi)=T_1 \quad \text{for } \xi > 0 \quad (10a)$$

$$f(\xi)=T_0 \quad \text{for } \xi < 0 \quad (10b)$$

This means that there is a thermal front of radius R_t given by $\xi=0$

$$\gamma \rho_1 c_1 \frac{4}{3} \pi (a^3 - R_t^3) = \rho c \frac{4}{3} \pi (a^3 - R_t^3) \quad (11)$$

behind which the liquid and particle temperatures are equal to the injected hydrazine temperature T_1 , and ahead of it equal to the initial particle temperature T_0 . The thermal front goes always behind the liquid front, $R_t > R$, as can be seen from equation (11) (figure 2). Equation (11) expresses a heat balance between the heat convected by the liquid hydrazine inside the particle and the non-steady particle heating.

2.2 Thermal front structure

To study the flame front structure we shall assume it to be thin compared with the particle radius a , and make the following change of variables

$$r = R_t(t) + \delta x \quad (12)$$

where δ is a characteristic thickness of the front

$$\delta \ll a \quad (13)$$

and x a non-dimensional length of order unity. The time is made nondimensional with a quantity of the order of t_r

$$\tau = \frac{Q_0}{2\pi a^3} \frac{\rho_1 c_1}{\rho c} t \quad (14)$$

By making this change of variables in equation (3), it is obtained

$$\frac{\partial T}{\partial \tau} + \beta(\tau)x \frac{\partial T}{\partial x} = \frac{\partial^2 T}{\partial x^2} \quad (15)$$

after neglecting terms of order δ/a , and choosing δ of the form

$$\delta^2 = \frac{K_p}{\rho_1 c_1} \frac{2\pi a^3}{Q_0} = \frac{3}{2} \frac{t_r}{t_c} a^2 \quad (16)$$

and

$$\beta = \frac{Q}{Q_0} \left(\frac{a}{R_t} \right)^3 \quad (17)$$

and using equations (11) and (2) for the thermal front radius R_t equation (16) expresses that condition (13) is satisfied if t_r is much smaller than t_c (as given by equation (5)).

The appropriate boundary and initial conditions for equation (15), if condition (13) is satisfied, are

$$x \rightarrow \infty \quad T = T_0 \quad (18a)$$

$$x \rightarrow -\infty \quad T = T_1 \quad (18b)$$

$$t = 0 \quad x < 0 \quad T = T_0 \quad (19)$$

Equation (15) and conditions (18) and (19) admit a similarity solution of the form

$$T = T_1 + \frac{T_0 - T_1}{2} \operatorname{erfc} z \quad (20)$$

Where

$$z = \frac{x}{\phi(\tau)} \quad (21)$$

and $\phi(\tau)$ is a function of time satisfying a differential equation of the form

$$\phi \dot{\phi} - \beta \phi^2 - 1 = 0 \quad (22)$$

with the initial condition

$$\tau = 0 \quad \phi = 0 \quad (23)$$

and β a function of τ given by

$$\beta = \frac{Q}{Q_0} \left(-\frac{a}{R_t} \right)^3 \quad (24)$$

The solution of (22) and (23) is given by

$$\phi^2 = 2 \exp \left(\int_0^\tau 2\beta d\tau \right) \int_0^\tau \exp \left(-\int_0^\tau 2\beta d\tau \right) d\tau \quad (25)$$

and using the expression (17) for β and equations (2) and (11) for R_t , it is obtained for ϕ

$$\phi^2 = 2 \left(-\frac{a}{R_t} \right)^4 \int_0^\tau \left(-\frac{R_t}{a} \right)^4 d\tau \quad (26)$$

The thermal front has a thickness that increases with time according to the law given by equation (26)

2.3 Conclusions and Discussion

The present theory indicates that when there is a situation for which condition (13) is satisfied the appropriate temperature to take into consideration to calculate the liquid residence time within the catalyst particle is the initial particle temperature, and the liquid hydrazine temperature does not have influence on this starting period. As the liquid residence time increases the thickness of the thermal front increases and may become of the order of the particle radius. This limiting situation arises when

$$\delta \sim a, \quad \frac{\delta c a^2}{K_p t_r} \sim 1 \quad (27)$$

An analytical expression for t_r is given in ref. 8 in which it is shown that t_r behaves with particle radius and temperature in the form

$$t_r = \frac{a}{T} \exp\left(\frac{5809}{T}\right), \quad T \text{ in } ^\circ\text{K} \quad (28)$$

showing that this criteria is particularly sensitive to temperature variations. For initial particle temperatures below 290°K and particle radius of 0.5mm. t_r becomes also dependent on liquid hydrazine temperature, according to the results of refs. 8, 9, that give then for t_r values above .1sec.

A different situation to be discussed is also what happens for larger values of T when t_r becomes smaller of the order of the millisecond for $a=0.5\text{mm.}$;

then the results of refs. 8,9 show that the liquid penetrates a small depth, "e", inside the particle and criteria (27) should be substituted by

$$\frac{\delta c e^2}{K_p t_r} \sim 1 \quad (29)$$

However, an asymptotic analysis carried out in ref.8 shows that "e" decreases precisely like $t_r^{-1/2}$, so that both effects compensate and the above presented theory is expected to be valid in this limit.

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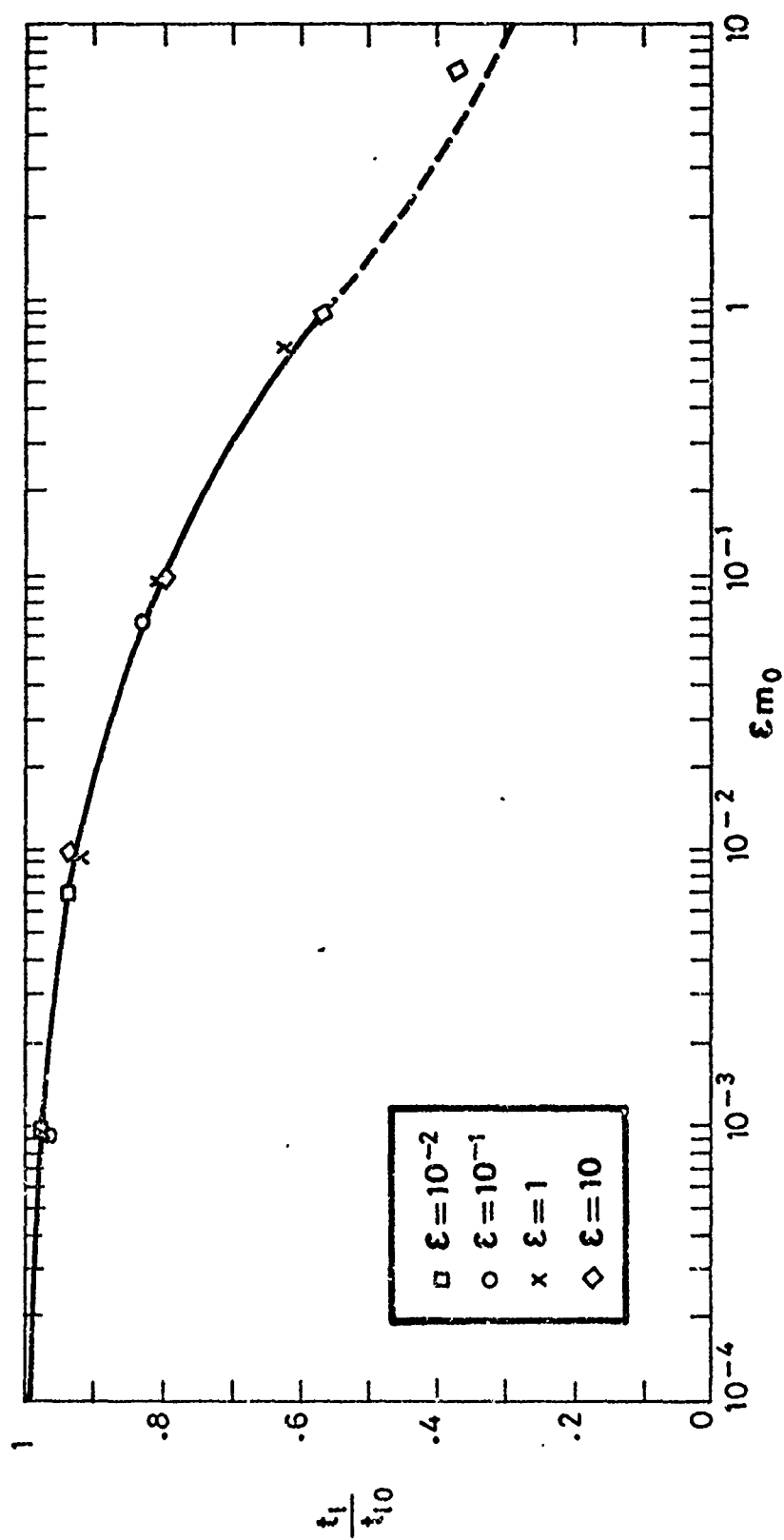
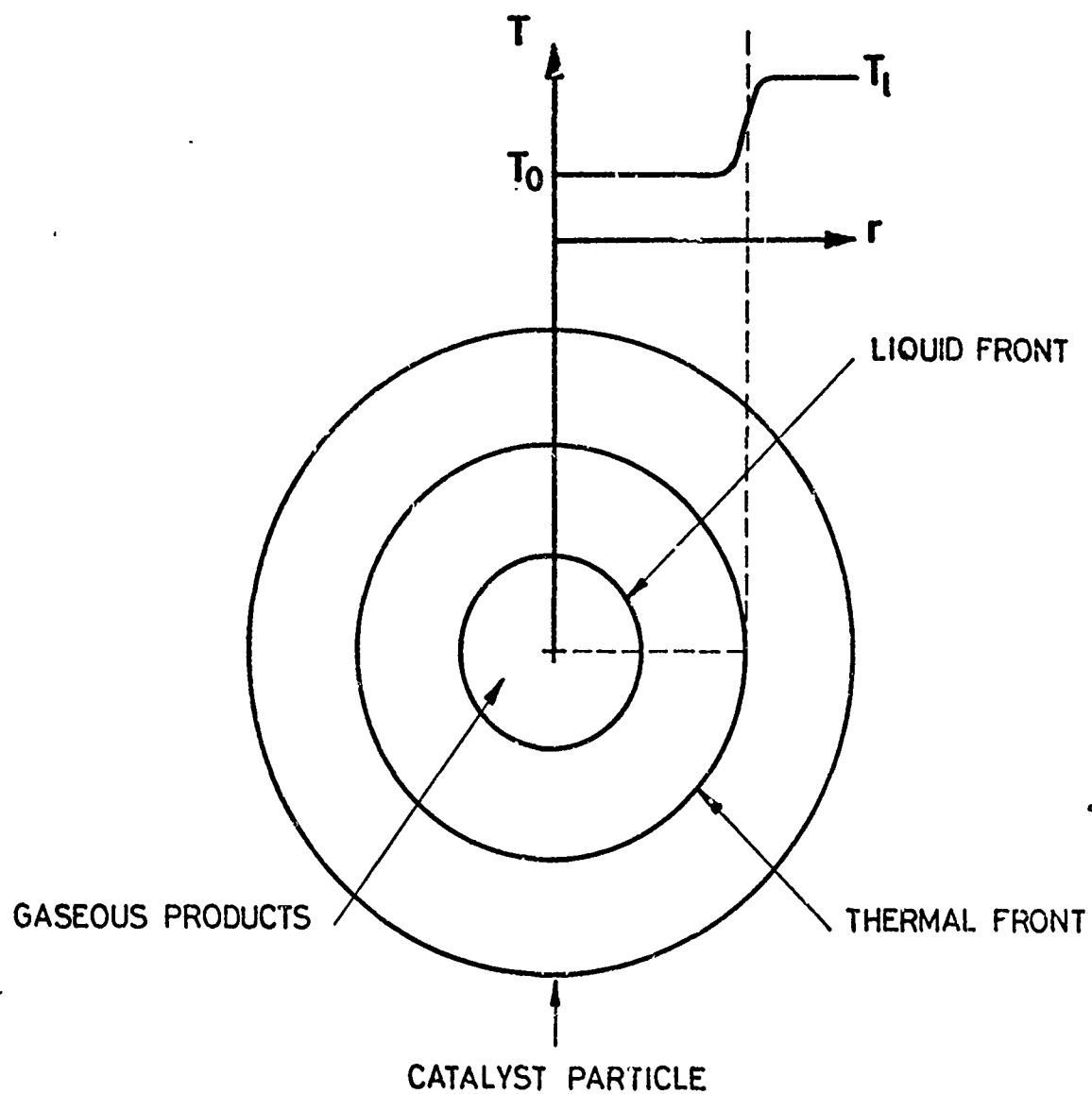


Fig.1.- Vaporization time as a function of ϵm_0 . Comparison with numerical sphere dissolution lifetime results of Duda and Vrentas¹.



Schematic showing temperature distribution in the particle.

Figure 2